

Critical Current and Topology of the Supercooled Vortex State in NbSe₂

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We study the behavior of the critical current, $I_c(H, T)$, of pure and Fe doped NbSe₂ crystals in the denominated disordered vortex region, limited by the critical field $H_{c2}(T)$ and the field $H_p(T)$ at which the peak effect in $I_c(H, T)$ is detected. The critical current follows an individual pinning response as demonstrated by its field independent universal function of the superfluid density. Transport measurements combined with Bitter decorations show no evidence of the existence of an amorphous phase in the high temperature region.

I. INTRODUCTION

Although the peak effect in the critical current, $I_c(H, T)$, at a field $H_p(T)$ close to the upper critical field, $H_{c2}(T)$, has been observed years ago its origin is still subject of debate. The traditional interpretation of this effect is based on the explanation suggested by Pippard¹ and on the formal treatment made by Larkin and Ovchinnikov (LO)². In this scenario the peak effect appears as a response of the vortex lattice to the presence of quenched disorder. The peak occurs due to the softening of the elastic constants in a crossover from local to non-local elasticity when approaching $H_{c2}(T)$. Using this formalism the behavior of $I_c(H, T)$ at the peak in NbSe₂ has been explained.³

The elastic description of the peak effect has been criticized by other authors⁴ emphasizing the necessary dynamic aspects of the phenomenon. An extensive analysis through transport measurements in NbSe₂ showed⁴ the plastic fingerprint predicted in 2D simulations^{5,6} in the $H-T$ region where the peak effect appears. In Ref. 4 a phase diagram with a liquid vortex phase for $H > H_p(T)$ is proposed.

Recent electrical transport measurements using Corbino-contact configuration have suggested⁷ other interpretation of the peak effect phenomenon. An abrupt decrease of $I_c(H, T)$ at $H_p(T)$ is considered to be the manifestation of a genuine first order thermodynamic phase transition from a disordered ($H > H_p(T)$) to an ordered vortex state ($H < H_p(T)$). In this scenario the width of the transition detected using the conventional four contact configuration is due⁸ to the injection of a disordered vortex phase through the surface. It is widely accepted^{9,10} that the critical current is history dependent for $H < H_p(T)$ while for $H_p(T) < H < H_{c2}(T)$ it is reversible. A dramatic example of history dependence of $I-V$ curves has been recently reported¹⁰ using a fast transport measurement technique. This technique allows to define a large critical current when the sample is cooled, in the presence of a field (FC), down to temperatures well below that of the peak in $I_c(H, T)$. On the other hand, when using a slow transport technique a

low critical current is detected. When the field is applied after zero field cooling the sample (ZFC) to temperatures below that of the peak in the critical current, the same low value of $I_c(H, T)$ is obtained independently of the technique, fast or slow, used to measure it.

Within the frame of this work the most relevant proposed properties of the mixed state associated with the presence of the peak effect can be summarized as follows: (a) The vortex phase nucleates at $H_{c2}(T)$ as a topologically disordered structure with a large critical current that remains in thermodynamic equilibrium, down to the temperature where the peak in $I_c(H, T)$ takes place; (b) when the sample is FC to lower temperatures the disordered vortex structure is supercooled as a metastable state with a large $I_c(H, T)$; (c) the thermodynamic equilibrium state obtained by ZFC experiments at low temperatures corresponds to a topologically ordered phase with low critical current.

Independently of the different interpretations of the peak effect there is consensus on the existence of an equilibrium disordered vortex state at temperatures above that of the peak, where transport measurements show no history dependence. It has been claimed^{7,9} that the disordered equilibrium phase shows a reentrance at fields low enough. In particular, in Fe doped samples the reentrant region has been found a field of 1000 Oe.⁷

The history independence of the transport properties when $H > H_p(T)$ suggests that the vortex structure is displaced homogeneously through the sample when the measuring current exceeds the critical one. However, the analysis of the field and temperature dependence of $I_c(H, T)$ in the so called disordered phase is still lacking. On the other hand, the claim that in FC experiments the disordered phase remains in a metastable state at low temperatures provides us the possibility of using the Bitter decoration to visualize its topology. This is particularly important because despite the claim of the presence of a disordered phase based on the analysis of $I_c(H, T)$ measurements, there is no trivial relation¹¹⁻¹⁵ between critical current and topological order. The critical current is determined by vortex displacements in the small length scale associated with the range of forces of

the pinning potential (superconducting coherence length $\xi(T)$), while the vortex topology responds to a larger length scale (lattice parameter), see Ref. 14.

In this work we focus the attention on the study of the field and temperature dependence of the critical current and its relation with the corresponding vortex structure of the proposed disordered phase. This is accomplished by complementing the transport measurements with Bitter decoration of the pristine FC vortex structure before any external force is applied. Thus, the Bitter image depicts the vortex structure tested by fast transport measurements.

II. LARKIN-OVCHINNIKOV THEORETICAL BACKGROUND

Since the LO theory has been shown³ to be appropriate to provide a quantitative understanding of $I_c(H, T)$ in the peak effect, we take the theoretical prediction of the model as the basic ingredient for the analysis of $I_c(H, T)$.

This model predicts an enhancement of the critical current when approaching $H_{c2}(T)$ due to the softening of the vortex lattice associated with the dispersive character of the tilt modulus. The increase of J_c is associated with a monotonous reduction of the LO correlation volume $V_c = R_c^2 L_c$, where R_c is the elastic correlation length perpendicular to H and L_c is the parallel one. At $H_p(T)$, R_c takes the minimum value compatible with the existence of a finite density of vortices, $R_c = a_0$, where $a_0 = 1.075(\Phi_0/B)^{1/2}$ is the vortex lattice constant. In this picture it is assumed³ $R_c = a_0$ in the whole region of fields $H_p(T) < H < H_{c2}(T)$ or equivalently $T_p(H) < T < T_{c2}(H)$.

In the LO theory the critical force is given by $BJ_c V_c = fN^{1/2}$, where N is the effective number of pinning centers in V_c and the pinning force, f , is usually assumed to be proportional to Ψ^2 , where Ψ is the Ginzburg-Landau order parameter. Close to $H_{c2}(T)$ the condensation energy decreases as $\Psi^2 \propto (1-b)(1-t)$ where $t = T/T_c$ and $b = B/H_{c2}(T)$. It is easily seen that $(1-b)(1-t) = \Theta/T_c$ where $\Theta = T - T_{c2}(H)$. Within these considerations and using the non-local elastic constant $C_{44}^* = C_{44}(1-b)(1-t)/\kappa^2$, where $C_{44} \approx H^2$ is the local tilting elastic constant¹⁶, it is found² that for $H > H_p(T)$

$$V_c = L_c a_0^2 = a_0^2 (4C_{44}^* a_0^2 / f n^{1/2})^{2/3} \propto H^{-1/3} \quad (1)$$

where it has been assumed that close to $H_{c2}(T)$ all pinning centers in V_c are effective: $N = nV_c$. Thus, the field and temperature dependence of the critical current is

$$J_c(H, T) \propto H^{-5/6} \Theta \quad (2)$$

Most of the experiments reported in the literature have been made at constant temperature varying field. In this case the theory predicts that the critical current decreases as $(1-b)$ and the rapid change of $J_c(H, T)$ as a function of b has been associated to this field dependence. However,

this behavior is difficult to check due to the narrow available experimental field range between $H_p(T)$ and $H_{c2}(T)$.

In order to verify the behavior predicted by Eq. 2 we investigated both, the field as well as the temperature dependence of $J_c(H, T)$. It should be noticed that Eq. 2 has an explicit dependence in magnetic field when $J_c(H, T)$ is plotted as a function of Θ . This dependence should be evident when measuring the critical current as a function of temperature for different fields. In this way it can be proved whether the vortex structure corresponds to an homogeneous vortex phase described by a correlation volume limited in its transverse direction, $R_c = a_0$.

III. EXPERIMENTAL

The experiments were performed in pure and Fe doped NbSe₂ single crystals. Material parameters for the Fe doped sample are $T_c = 5.8$ K, $\delta T_c(10\% - 90\%) = 0.2$ K, $\xi(0) = 78$ Å and for the pure sample $T_c = 7.02$ K, $\delta T_c(10\% - 90\%) = 0.1$ K, $\xi(0) = 89$ Å.

The results for the pure and impure samples are qualitatively similar. However, measurements of the Fe doped sample have a relative advantage over the pure one due to the wider field region $H_p(T) < H < H_{c2}(T)$.

The transport properties were measured using the Corbino-contact configuration as shown in the insert of Fig. 6 (b). A dc current was injected between the central contact and the four contacts indicated in the figure. The distance between I^- and V^- is approximately 0.3 mm and between V^+ and V^- is 0.3 mm, in both cases the sample thickness is around 40 μ m.

The sample was thermally anchored to a sapphire glued to a copper sample holder that was thermally weakly connected to a liquid helium bath at 4.2 K. A heater on the sample holder was used to control the temperature of the sample by means of a PID controller.

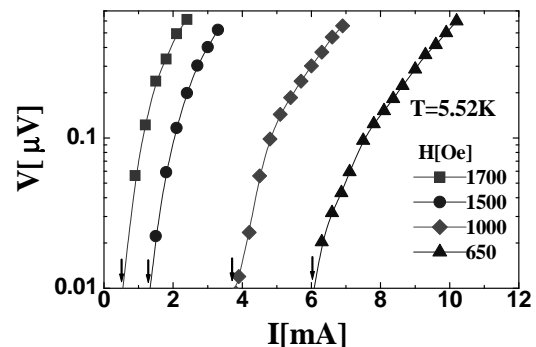


FIG. 1. Typical $I - V$ curves for the Fe doped sample using the Corbino-contact configuration, where the critical current is marked with an arrow. The measurements were made at different magnetic fields and $T = 5.52$ K greater than $T_p(H)$.

Measurements were made either cooling the sample in a field, FC experiments, or cooling the sample in zero field and then sweeping the field at a given temperature, ZFC.

The critical current was defined as that inducing a voltage of 10 nV in the Fe doped sample and of 20 nV in the pure sample. Moderate changes in voltage criteria to define $I_c(H, T)$ do not modify the conclusions reached in the paper.

The Bitter decoration in pure samples was performed at 4.2 K in FC experiments following the procedure described in Ref. 17. The magnetic decoration of the Fe doped sample was made at 3 K in order to achieve the necessary resolution. The ZFC decorations of pure samples were made applying the field after cooling the sample down to 4.2 K.

IV. RESULTS AND DISCUSSION

Figure 1 shows typical $I-V$ curves for different magne

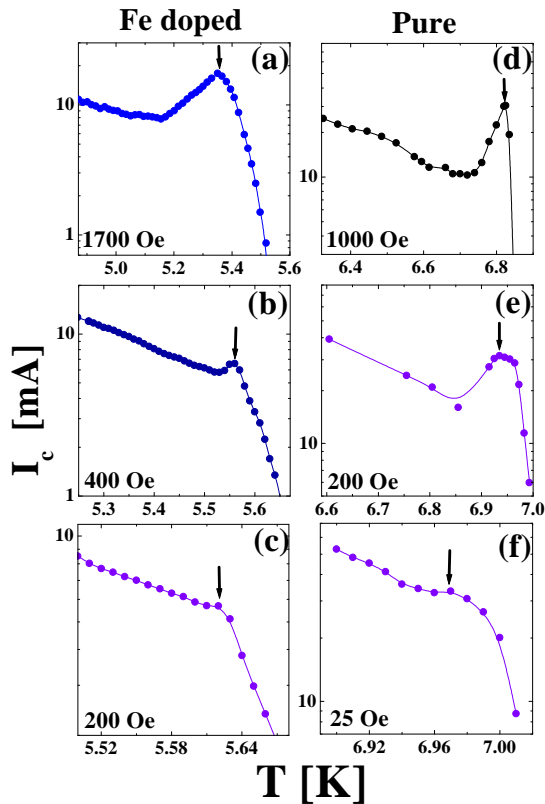


FIG. 2. Critical current curves for different magnetic fields as a function of temperature for the Fe doped sample: (a), (b), (c) and for the pure sample: (d), (e), (f). The position of the peak in $I_c(H, T)$ is indicated by an arrow. Figures (c) and (f) show the $I_c(H, T)$ curves at the lowest field where the peak is detected for each sample.

tic fields obtained in the Corbino contact configuration for the Fe doped sample in the temperature range $T > T_p(H)$. The critical current for each magnetic field is marked with an arrow. Similar results are obtained for the pure sample. For the same value of $I - I_c$ the voltage increases with H , as expected.

Critical current curves as a function of temperature for different magnetic fields are shown in Fig. 2 for Fe doped and pure NbSe₂ samples. A well defined peak in $I_c(H, T)$ is found for fields higher than 200 Oe and 25 Oe for the Fe doped and pure samples respectively (see Fig. 2(c) and (f)). Below those fields only a change in the slope of $I_c(H, T)$ is detected when approaching $T_{c2}(H)$. We have defined $H_{c2}(T)$ (or $T_{c2}(H)$) as the magnetic field (or temperature) where the critical current extrapolates to zero. The results are shown in Fig. 6 (a) and (b).

The critical current of both materials obtained with the described experimental setup is history independent and consequently $I_c(H, T)$ is a single-valued function of H and T . An example is shown in Fig. 3 for the Fe doped sample. Most of the data reported in this work was taken sweeping temperature.

To verify whether the LO theory (Eq. 2) describes the field and temperature dependence of the results shown in Fig. 2, the critical current for different fields is plotted as a function of the variable Θ , see Fig. 4. Contrary to what is expected from Eq. 2, the critical current in the variable Θ becomes field independent in a region of temperatures higher than that where the peak in $I_c(H, T)$ takes place : $-0.1 \leq \Theta \leq 0$ for the Fe doped sample and $-0.03 \leq \Theta \leq 0$ for the pure case. This is clearly shown in Fig. 5 where $I_c(H, \Theta)$ as a function of magnetic field for different values of Θ is plotted. This field independent regime for the critical current is obeyed even for the range of fields where the peak effect is not observed, see Fig. 4(a).

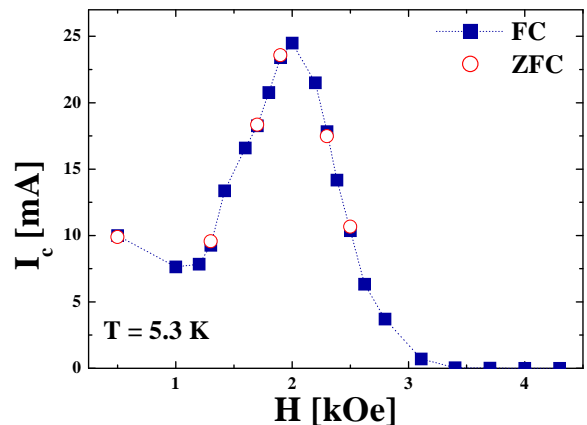


FIG. 3. Field dependence of $I_c(H)$ at $T = 5.3$ K in the Fe doped sample. The filled square symbols depict the peak effect for FC measurements while the open circles are for the ZFC case.

As a consequence of the result described in the previous paragraph it is concluded that the LO theory (see Eq.2) is not applicable in the Θ region where the critical current is field independent. Besides this, the field dependence of $I_c(H, \Theta)$ at the peak does not follow the behavior predicted by the theory in the limit $R_c = a_0$. The predicted increase of $I_c(H, \Theta)$ at the peak when decreasing the magnetic field is not observed in any of the samples. Thus, the behavior of the critical current in the peak effect region is not qualitatively described by the LO collective theory under the assumption $R_c = a_0$.

The results in Figs. 4 and 5 make evident that the field and temperature dependence of the critical current are only determined by the change of superfluid density as described by $\lambda_h^{-2}(T, H) \propto \Psi^2 \propto (1-t)(1-b) = \Theta T_c$, when approaching $T_{c2}(H)$. The lack of explicit field dependence of $I_c(H, \Theta)$ indicates the irrelevance of the vortex-vortex interaction to determine the pinning energy, suggesting that the critical current is fully determined by the pinning of individual vortices: single vortex limit. This seems to be a counter-intuitive and surprising result in a field region where the distance between cores is smaller than the interaction characteristic length,

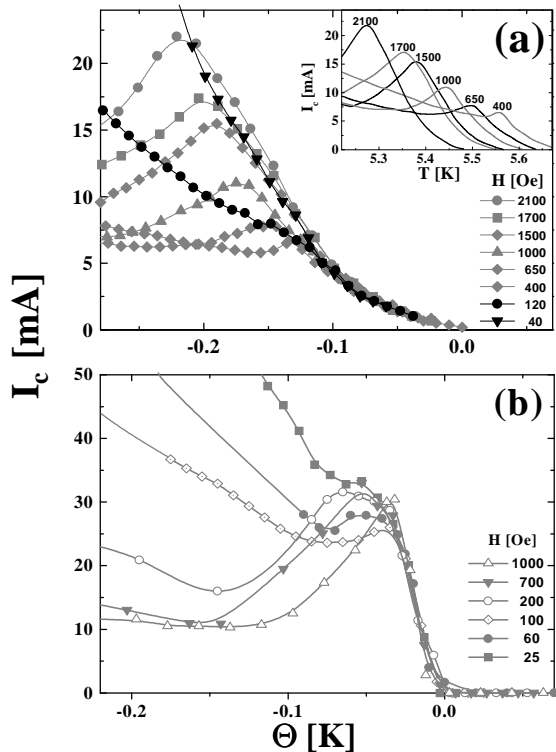


FIG. 4. Critical current as a function of the temperature Θ (see text) for different magnetic fields. (a) Fe doped sample: Dark symbols correspond to the results where no peak in $I_c(H, \Theta)$ is detected. The insert shows the same curves as a function of the absolute temperature. (b) Pure sample. The data shows no agreement with the LO theory, see text.

$\lambda_h(T, H)$. We will come back to this point in the conclusions.

Figure 6 summarizes the results described up to now in a B - T phase diagram. In Fig. 6 (a) the line $H_p(T)$ indicates the field where the peak in $I_c(H, T)$ takes place and the line $H_s(T)$ marks the crossover to the single vortex regime (lack of field dependence of $I_c(H, \Theta)$) for the Fe doped sample. The Fig. 6 (b) shows the results for the pure sample, in this case $H_s(T)$ coincides with $H_p(T)$. The new individual vortex regime line $H_s(T)$ is found to follow the condition $\Theta_s = T_s(H) - T_{c2}(H) = \text{constant}$.

In order to detect a possible reentrance of the line $H_p(T)$ in the phase diagram of the Fe doped sample as reported in Ref. 7, we have made careful measurements of critical current as a function of temperature or magnetic field keeping constant field or temperature respectively. No evidence of a reentrant $H_p(T)$ was found for the investigated sample (the paths followed to detect the possible reentrance are indicated with dotted lines in Fig. 6(a)). The presence of a peak effect down to 200 Oe and the absence of reentrance in the Fe doped sample is a different behavior than that found for similar samples in Ref. 7 (comparable T_c and $\xi(0)$) where the reentrance of the high temperature phase takes place at $H = 1000$ Oe.

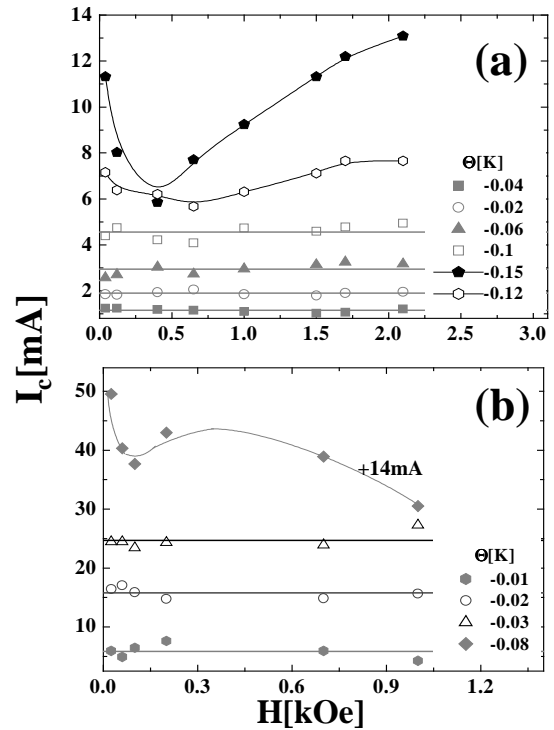


FIG. 5. Plot of $I_c(H, \Theta)$ as a function of H for different Θ . For the Fe doped sample (a) the single pinning regime is found for values of $\Theta \geq -0.1$ and for the pure sample (b) for $\Theta \geq -0.03$.

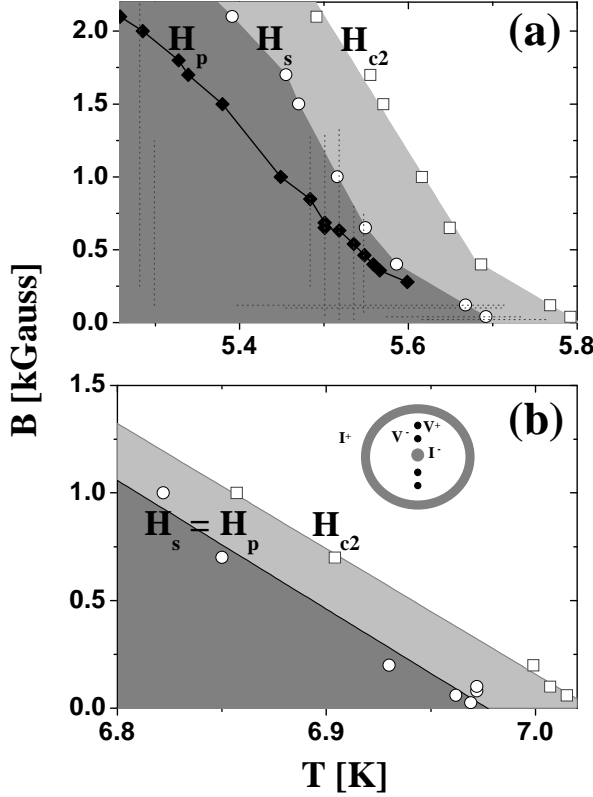


FIG. 6. $B-T$ phase diagrams. (a) Fe doped sample ($T_c = 5.8$ K). $H_{c2}(T)$ is the field where I_c goes to zero. The $H_p(T)$ line is the position of the peak of $I_c(T, H)$. The line $H_s(T)$ corresponds to a crossover to a single pinning regime. Dotted lines indicate the paths followed to study the reentrance of the disordered vortex phase. (b) Pure sample ($T_c = 7.02$ K). In this case the $H_s(T)$ line coincides with $H_p(T)$. In the insert is shown the Corbino contact configuration.

This clearly shows that the reentrant peak effect depends strongly on unknown properties of the sample and consequently on the ubiquity of the supposed disordered phase as has been verified by several authors.¹⁸

The results of magnetic decorations performed in FC experiments at 60 Oe are shown in Fig. 7(a) for the pure sample at 4.2 K and in (b) for Fe doped sample at 3 K. It is important to remark that in this case the FC decoration for the Fe doped sample is made at a field below the lowest field where the peak is detected. In the pure case the FC at 60 Oe crosses the $H_s(T)$ line. An identical polycrystalline nature of the FC state is observed in both type of materials. Following Ref. 10, this structure corresponds to the so called metastable disordered phase. The polycrystalline structure is detected independently of the presence of the peak effect at the field where decoration is made. This indicates that the FC polycrystalline structure is originated in the region of temperatures where the single vortex regime dominates. This is an interesting result showing that some degree

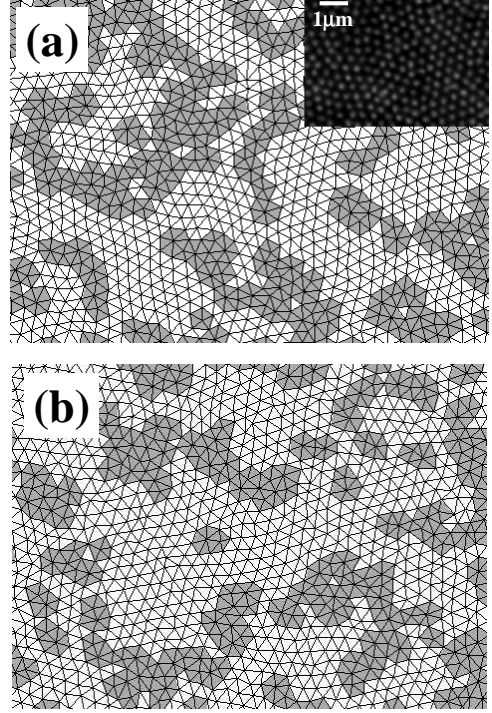


FIG. 7. FC vortex structure in NbSe₂ observed by magnetic decoration at 60 Oe. Delaunay triangulations where the nearest neighbors vortices are bonded and the non-sixfold coordinated are marked in gray. (a) Pure sample decorated at 4.2 K and (b) Fe doped sample decorated at 3 K.

of orientational order is preserved when nucleating the vortex structure at $H_{c2}(T)$ in a single vortex limit.

The Bitter structure obtained in ZFC experiments at 4.2 K shows gradients of B as a result of the presence of transport currents induced when applying the field.¹⁷ Despite this, a polycrystalline structure is found in most of the sample. The average size of the grains coincides with those shown in Fig. 7 obtained in FC experiments.

V. CONCLUSIONS

We have found a universal behavior of the temperature and field dependent critical current when approaching the mean field superconducting transition, $T_{c2}(H)$. The universal character is made evident when plotting $I_c(H, T)$ as a function of the temperature difference $\Theta = T - T_{c2}(H)$, as shown in Figs. 4 and 5. This shows that the superfluid density univocally determines the pinning properties of the material in this region of the phase diagram. The lack of explicit field dependence of $I_c(H, \Theta)$ when plotted as a function of Θ indicates that the vortex-vortex interaction plays no significant role in the determination of vortex pinning. The results cannot be inter-

preted within the collective pinning theory in the single vortex limit² since the distance between vortex cores is much smaller than the effective penetration depth, λ_h . This rather suggests that in this large λ_h limit, the interaction of individual pinning centers with a vortex line overcomes the weak interaction between vortices.¹⁹

It is important to remark that the novel single pinning regime is not necessarily associated with the presence of the peak effect: despite the peak effect is not detected below 200 Oe in the Fe doped sample the crossover to single pinning takes place even at lower fields.

The transport and Bitter experiments make evident that the single vortex pinning found at high temperatures is compatible with a structure where directional order is preserved within the grains. This reinforces the claim that the typical length scale associated with critical current is different from that characterizing the topological order.¹⁴

We have not been able to detect a reentrance of $H_p(T)$, neither in the Fe doped sample nor in the pure one. Whether this indicates that the phenomenon fades away when decreasing field or transforms into a weak signal not detected in our samples deserves further research.

The combined transport and Bitter decoration experiments together with the results in Ref. 10 open a question mark on the nature of the disordered phase. This is of particular importance in view of recent data interpreted^{7,20,21} in terms of an order-disorder thermodynamic transition that takes place at the temperature where the peak in $I_c(H, T)$ is observed. The results reported in this paper call for a better definition of what is meant by order and disorder characterized by critical current measurements. The topology of the FC structure as observed by Bitter decoration shows no gradient of B indicating the absence of transport currents acting on the bulk of the vortex system.^{22,23} Although this supports that by FC the high temperature structure is supercooled, the results make evident that the high temperature phase is not amorphous.

The observed ZFC vortex structure is not an ordered equilibrium state; it is consistent with a topological unstable state dominated by the presence of transport currents.¹⁷

In conclusion, through the analysis of the vortex structure and the assumption of the existence of the proposed supercooling of the high temperature phase in the FC experiments, no evidence of an order-disorder phase transition is found in samples where the transport properties show anomalies associated with a first order thermodynamic transition.⁷

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field and the magnetic induction is always less than 10%.

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